SURFACE PLASMON AMPLIFICATION BY STIMULATED EMISSION OF RADIATION

STATEMENT REGARDING RELATED APPLICATIONS

This application claims the benefit of priority to U.S. Provisional Application No. 60/437,760, entitled, "Surface Plasmon Amplification By Stimulated Emission of Radiation," filed January 3, 2003, the contents of which are hereby incorporated by reference.

FIELD OF THE INVENTION

The present invention relates to generating surface plasmons and more specifically relates to quantum generation of localized optical fields on a nanoscale.

BACKGROUND OF THE INVENTION

Electromagnetic radiation propagates as a wave and, in an isotropic uniform medium, consists of oscillating electric and magnetic fields at right angles to one another and to the propagation direction. Electromagnetic radiation comes in discrete packets known as photons. Photons are the basic unit of light. Photons were first postulated by Planck who showed that electromagnetic radiation had to come in discrete units. Because the energy of photons is directly proportional to their frequency, low-energy photons have low frequencies, while high-energy photons have high frequencies. Low-energy photons include radio waves or microwaves, medium-energy photons include visible light, high-energy photons include X-rays, while those having higher energy still are called gamma rays.

LASER is an acronym for "light amplification by stimulated emission of radiation." The first working laser was built in 1960 and made use of optical pumping of a ruby crystal from a flash lamp. The first continuous laser was produced

in 1962 using an arc lamp instead of a flash lamp. Since its development, the laser has been used in a multitude of different applications and has become virtually ubiquitous in consumer electronics. A laser emits an electromagnetic wave having electrical field and magnetic field components. Unfortunately, the light emitted by a laser is an electromagnetic wave that cannot be localized at target regions significantly smaller than the wavelength (on order of fractions of a micron for visible light). So, while lasers are an extraordinarily valuable technology, the use of lasers is generally limited to applications in which the target regions are significantly larger than the wavelength of the laser light. This has excluded the practical use of the laser from various biotechnical applications, for example, where the target region is on the order of a few nanometers (1 nanometer (nm) = 10^{-9} meter) and is thus 100 to 1000 times smaller than a typical laser wavelength.

There are several well-known devices and methods for channeling the energy of laser light to the nanoscale using surface plasmon resonances. One of them is apertureless NSOM (near-field scanning optical microscope), which uses a sharp metal (usually, gold or silver) tip with the radius of curvature of typically 30 to 50 nm, irradiated by an external laser light. This radiation excites surface plasmon oscillations at the tip, creating high oscillating local fields localized at the tip in nanoscale areas with sizes comparable to tip's curvature. These localized oscillating electric fields are used to probe surfaces and molecules with resolutions on the order of approximately 30-100 nm. The limitation of such devices is that the only a negligible (typically, 10^{-7}) fraction of the laser energy is concentrated on nanoscale. It is, therefore, difficult or impossible to control properties (e.g., specific plasmon modes excited, shape of the localization region, and polarization and amplitude of the fields). It also is difficult to fabricate an effective nanometer tip -- conventionally, only one in 20 tips work satisfactorily.

Another group of devices and methods have been developed to exploit surface plasmons for the sensing of chemical and biological agents. Such a device normally includes an interface between a metal and dielectric medium that possesses surface

plasmon modes. These modes are excited by an external laser source to create oscillating electric fields at this interface. These fields excite molecules adsorbed at this interface, where the detection is done by either measuring absorption resonances of the excitation laser light, or by detecting Raman scattering from those adsorbates. The limitation of such methods and devices is that they generally are only usable with a comparatively large number of molecules, are incapable of detection single molecules or biological particles, and they do not have nanometer-scale spatial resolution in the lateral direction.

Therefore, there is a need in the art for the generation of an oscillating electric field on a nanoscale (i.e., on the order of 1-100 nanometers). This electric field should result from the emission of surface plasmons (electric oscillations in matter) and should be able to be localized on a target region on the order of nanometers, i.e., by a factor of thousand shorter than the practical target region for a laser.

SUMMARY OF THE INVENTION

A nanostructure is used to generate a highly localized nanoscale optical field. The field is excited using surface plasmon amplification by stimulated emission of radiation (SPASER). The SPASER radiation consists of surface plasmons that undergo stimulated emission, but in contrast to photons can be localized within a nanoscale region. A SPASER can incorporate an active medium formed by two-level emitters, excited by an energy source, such as an optical, electrical, or chemical energy source. The active medium may be quantum dots, which transfer excitation energy by radiationless transitions to a resonant nanosystem that can play the same role as a laser cavity in a conventional laser. The transitions are stimulated by the surface plasmons in the nanostructure, causing the buildup of a macroscopic number of surface plasmons in a single mode. The SPASER may not emit light waves, because its two-level emitters undergo radiationless transitions where their excitation

energy is transformed into the quasi-static electric field energy of surface plasmons of the nanostructure. There are a multitude of possible applications of the SPASER in nanoscience and nanotechnology, including for near-field, in particular non-linear, optical probing and nanomodification.

The various aspects of the present invention may be more clearly understood and appreciated from a review of the following detailed description of the disclosed embodiments and by reference to the drawings and claims.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1a is a block diagram illustrating the primary components of an exemplary embodiment of the present invention.

Figure 1b is a block diagram illustrating the primary components of an alternative embodiment of the present invention.

Figure 2 is a block diagram depicting the application of energy to the active medium in an exemplary embodiment of the present invention.

Figure 3 is a block diagram depicting a highly localized, nanoscale, oscillating electric field that is generated in response to the application of energy to the active medium in an exemplary embodiment of the present invention.

Figure 4 is a flow chart depicting a method for building a SPASER device that is an exemplary embodiment of the present invention.

Figure 5 is a flow chart depicting a method for generating a highly localized nanoscale electric field that is an exemplary embodiment of the present invention.

Figure 6 is a block diagram depicting an exemplary spectroscopy application that is an exemplary embodiment of the present invention.

Figure 7 is a block diagram depicting an exemplary electronic data storage and retrieval application that is an exemplary embodiment of the present invention.

Figure 8 is a block diagram of a pseudo-SPASER device that generates both localized electrical and magnetic fields that is an alternative embodiment of the present invention.

DETAILED DESCRIPTION OF THE EXEMPLARY EMBODIMENTS

In exemplary embodiments of the present invention, a nanostructure is used to generate a highly localized nanoscale optical field. The optical field is excited using Surface Plasmon Amplification by Stimulated Emission of Radiation (SPASER). The SPASER radiation consists of surface plasmons that undergo stimulated emission, but in contrast to photons can be localized within a nanoscale region. A SPASER can incorporate an active medium formed by two-level emitters, excited by an energy source, such as an optical, electrical, or chemical energy source. The active medium may be quantum dots, which transfer excitation energy by radiationless transitions to a resonant nanosystem that can play the same role as a laser cavity in a conventional laser. The transitions are stimulated by the surface plasmons in the nanostructure, causing the buildup of a macroscopic number of surface plasmons in a single mode.

The SPASER may not emit light waves, because its two-level emitters undergo radiationless transitions where their excitation energy is transformed into the quasi-static electric field energy of surface plasmons. There are a multitude of possible applications of the SPASER in nanoscience and nanotechnology, including for near-field non-linear optical probing and nanomodification.

A detailed summary of the theoretical aspects of the present invention are described in a paper authored by the inventors: D. J. Bergman and M. I. Stockman, *Surface Plasmon Amplification by Stimulated Emission of Radiation: Quantum Generation of Coherent Surface Plasmons in Nanosystems,* Phys. Rev. Lett. 90, 027402 (2003). That paper is hereby incorporated by reference. The contents of that

paper were included in U.S. Provisional Application No. 60/437,760, filed on January 3, 2003 and to which this patent application claims priority.

Figure 1a is a block diagram illustrating the primary components to an exemplary embodiment of the present invention. In the embodiment of Figure 1a, a SPASER device is depicted. Spaser is an acronym for surface plasmon amplification by stimulated emission of radiation. The SPASER device of Figure 1a includes a resonant medium **100** that is located adjacent to an active medium **102**. The active medium **102** is contained within a carrier medium **104**. In the embodiment of Figure 1a, the active medium **102** and the resonant medium are affixed adjacent to one another to a substrate **106**.

In this embodiment of the present invention, the active medium **102** is placed adjacent and on top of the resonant medium **100**. The resonant medium **100** is affixed to the substrate **106**. The active medium **102** and/or the carrier medium **104** can be affixed to the resonant medium **100** or to the substrate **106**. The active medium **102** may cover substantially the entire resonant medium **100**, such that the resonant medium is substantially surrounded by the active medium **102**; such a configuration is favorable for excitation of a broad range of surface-plasmon modes in the resonant medium. Alternatively, the active medium **102** may cover less than the entire resonant medium **100** for the purpose of mode selection: those surface-plasmon modes of the resonant medium with preferably be excited whose maxima are within the areas covered by the resonant medium.

The resonant medium may consist of a metal or composite (metal/semi-conductor/dielectric) nanoparticle that has surface plasmon modes in the ultraviolet, visible, or infrared region of the electromagnetic frequency spectrum. For example, the resonant medium **100** may be a silver nanosphere with a radius of between 10 and 50 nanometers (nm). Alternatively, the resonant medium may be a silver nanoshell with a thickness of between 5 and 20 nm and a radius of 50 nm. Those skilled in the art will appreciate that the resonant medium may be fabricated from various materials and of various sizes. Preferably, the resonant medium **100** will be fabricated of a

metal or composite nanoparticle having low dielectric losses at the generation frequency. This characteristic has been observed by the inventors in nanoparticles of so-called coinage metals, including silver, platinum, and gold, in alkaline metals, and in aluminum.

The resonant medium **100** depicted in Figure 1a is a nanoparticle in the form of a nanowedge. The nanowedge has a V-shaped form. The resonant medium nanoparticle **100** of Figure 1a is approximately 30 nm in length (i.e., along its longitudinal axis from the vertex to the opening) and approximately 20 nm in width. Preferably, the resonant medium **100** is a material that supports (i.e., possesses) surface plasmons. Although Figure 1a depicts the resonant medium **100** affixed to a substrate **106**, those skilled in the art will appreciate that the resonant medium nanoparticle may be either free-standing or deposited on a substrate **106**.

A second primary component of a SPASER device is the active medium **102**. The active medium may contain objects having significant dipole oscillator strength for transitions at a desired frequency. Examples of such objects include rare-earth ions and semiconductor quantum dots. Those skilled in the art will appreciate that other known objects exhibit this characteristic. The transition frequency preferably overlaps with the spectral frequency range of surface plasmons (e.g., from ultraviolet frequencies to infrared frequencies). Free particles exhibiting such transitions also exhibit efficient fluorescence. The active medium **102** is a collection of fluorescent (i.e., chromophoric) particles. Preferably, the fluorescent particles of the active medium **102** are quantum dots. The active medium **102** depicted in the exemplary embodiment of Figure 1a is embedded within a carrier medium **104**. The combination of the active medium **102** and the carrier medium **104** comprises a nanosize layer that can be affixed to a substrate **106** and/or to the resonant medium **100**. The shape of the active medium **102** can be designed to complement the size and structure of the resonant medium **100** such that the desired eigenmodes are generated.

The active medium **102** preferably comprises quantum dots. Quantum dots transfer their excitation energy by radiationless transition to a resonant nanostructure that fulfills the same role as a laser cavity in a conventional laser. The radiationless transitions are stimulated by the surface plasmons in the nanostructure, causing a buildup of a macroscopic number of surface plasmons in a single mode. Quantum dots are preferable, because of their desirable physical and chemical properties. Quantum dots are tunable in frequency due to quantum confinement. Quantum dots also have relatively large transitions dipoles and narrow transition lines and allow dense packing without compromising their optical properties. Quantum dots are well-known to be physically, chemically, and photochemically stable. Presently, there are two types of well-known quantum dots: nanocrystals grown chemically or quantum dots created by modification (including doping) of semiconductor surfaces. A surface class of nanocrystal quantum dots for the SPASER active medium is comprised of nanocrystals covered by a layer of organic molecules, which makes them even more chemically and mechanically stable.

The active medium **102** can be excited by an external energy source. Examples of external energy sources include optical energy sources, electrical energy sources and chemical energy sources. Although not likely to be feasible on the nanoscale, a nuclear energy source may also be used to excite the active medium **102**. For optical excitation, the excitation frequency is different than the generated frequency of the SPASER nanostructure. When excited, the active medium **102** undergoes a resonant transition, whereby energy is transferred to surface plasmons in the resonant medium **100**, as a result of electric interaction. The SPASER generates a localized radiation field due to the stimulated emission of surface plasmons.

As mentioned above, the stimulated emission of photons is a well-known phenomenon wherein the probability of a photon's emission in a certain state is proportional to the number of photons that are already present in that state. This is the phenomenon by which lasers sustain generation in a lasing mode. A similar theory applies to the stimulated emission of surface plasmons in a SPASER. Similar

to a laser, a SPASER has the ability to concentrate energy in time. The SPASER stores energy during a comparatively long period and may emit the energy as an ultra short pulse. The pulse of a SPASER can be on the order of **100** fs. The SPASER generates optical fields defined to be nanoscale (sizes on the order of 1 to **100** nanometers). In near-field scanning optical microscopes, light is generated outside of the nanoscale range and channeled into the nanoscale range. The SPASER is significantly different in that it generates optical fields on the nanoscale and does not need channeling. Notably, the SPASER does not necessarily emit light. That is, a purely electric oscillating field is associated with plasmon emission and there is no significant magnetic field component in the localized field. In general, the SPASER is useful in applications where light is not needed, but there is a need for localized optical field generation. Applications for the SPASER are discussed in more detail in connection with Figures 6 and 7.

Figure 1b is a block diagram illustrating the primary components of an alternative embodiment of the present invention. As stated above in connection with Figure 1a, in certain cases it is preferable that the resonant medium **110** is surrounded by the active medium **112**. However, in the embodiment in Figure 1a, the active medium **102** is applied to the top of the resonant medium **100** and there is no active medium **102** under the resonant medium **100**. That is, there is no active medium **102** between the resonant medium and the substrate **106**. In the alternative embodiment of Figure 1b, a second carrier medium **120** including suspended active medium **116** is affixed to a substrate **122** before the resonant medium **110** is affixed. The second carrier medium **114**, including a second layer of active medium **112**, is applied on top of the resonant medium **110**. Accordingly, the resonant medium **110** is completely surrounded by or "sandwiched" in the active medium **112/116**.

As stated above, those skilled in the art will appreciate that the substrates **106**, **122** depicted in Figures 1a and 1b are optional and the SPASER device depicted in those figures can be implemented using a free-standing resonant medium. For the purposes of the description of the various embodiments of the present invention, the

combination of the active medium and the carrier medium will be collectively referred to as the active medium. Those skilled in the art will appreciate that the carrier medium can be an inert, non-functional component of the SPASER, used only to provide desired structural characteristics to the active medium. Alternatively, the carrier medium may be completely absent if the aggregate of the emitters, e.g., quantum dots, is mechanically sufficiently stable. Yet another variation of the SPASER design may be that shown in Figure 1b where the upper active-medium layer **114** is absent. In such a case, the layer **120** is deposited on the substrate **122**, and the resonant medium **110** is affixed on top of it. Such a configuration can be advantageous for applications where the exposed resonant medium should be in the direct contact with or in close proximity to an external object. (e.g., a molecule or biological object for diagnostic purposes.)

Figure 2 is a block diagram depicting the application of energy to the active medium in an exemplary embodiment of the present invention. The SPASER **200** of Figure 2 comprises a resonant medium **206** and an active medium **204** deposited on a substrate **202**. SPASER generation can be accomplished by applying energy to the active medium **204**. An energy source **208** is used to apply energy to the active medium **204**, thus exciting the active medium **204** and causing the emission of surface plasmons in the resonant medium **206**. The energy source may be optical, electrical, chemical, or nuclear. The inventors contemplate that virtually any energy source that can excite the active medium can be used. In the case of an electrical energy source, nanoleads may be used to connect the active medium **204** to the electrical energy source. The nanoleads are not depicted in Figure 2.

Figure 3 is a block diagram depicting a highly localized nanoscale electric field that is generated in response to the application of energy to an active medium **304** in an exemplary embodiment of the present invention. The SPASER device **300** of Figure 3 generates a localized field response **302** when energy is applied to the active medium **304**. The response **302** is generated at the apex of the nanowedge resonant medium **306** and is localized in that a high magnitude oscillating electric

fields are located at or near the nanowedge vertex with significantly lower magnitude fields **308** in the near vicinity. Consequently, the SPASER device **300** can be used to apply a concentrated field to a target by placing the target in the vicinity of the localized field response **302**. Advantageously, the magnitude of the localized field response **302** can be adjusted. The adjustability of the localized field response **302** is one characteristic of the SPASER device **300** that makes it useful for applications requiring fields of various energy levels. For example, a higher magnitude energy field may be used for writing data onto a storage medium than for reading data from the storage medium.

Figure 4 is a flow chart depicting a method for building a SPASER device that is an exemplary embodiment of the present invention. The method of Figure 4 begins at start block **400** and proceeds to step **402**. At step **402**, the resonant medium is affixed to a substrate. As described above in connection with Figures 1a through 3, the resonant medium may be affixed to a substrate or free standing. Accordingly, step **402** is optional.

The method proceeds from step **402** to step **404**. At step **404**, the active medium is suspended in a carrier medium. As described above, the carrier medium can be an inert material in that it provides only structural characteristics for supporting the active medium in a particular physical location. Alternatively, an active medium without a carrier medium, e.g., dry aggregate of quantum dots, also can be used. In such a case, step **404** is optional.

The method proceeds from step **404** to step **406**. At step **406**, the carrier medium is affixed to the resonant medium. As described in connection with Figure 1b, the carrier medium (containing the active medium) can be affixed to the resonant medium, to the substrate, or to another carrier medium in the case where the resonant medium is surrounded by the carrier medium/active medium. The method of Figure 4 proceeds from **406** to step **408**.

At step **408**, a nanolead is connected between an energy source and the active medium. As stated above, the nanolead is used for applications of the SPASER

device wherein the active medium is excited using an electrical energy source. For applications wherein an energy source other than an electrical energy source is used, a nanolead may not be used and step **408** would then be optional. Those skilled in the art will appreciate that the nanolead may not be connected to an energy source upon the manufacture of the SPASER device, but may be connected only to the active medium such that the energy source may be connected upon use of the SPASER device. The method proceeds from step **408** to end block **410** and terminates.

Figure 5 is a flow chart depicting a method for generating a highly localized nanoscale electrical field that is an exemplary embodiment of the present invention. The method of Figure 5 begins at start block **500** and proceeds to step **502**. At step **502**, energy is applied to the active medium. As described above, conventional energy sources include optical, electrical, chemical, and nuclear energy sources. The method proceeds from step **502** to step **504**. At step **504**, the active medium is excited by the applied energy. The method then proceeds to step **506**, wherein the active medium undergoes resonant transition.

The method of Figure 5 proceeds from step **506** to step **508**. At step **508**, energy is transferred from the active medium to the surface plasmons in the resonant medium. The method then proceeds to step **510**, wherein the emission of surface plasmons in the resonant medium is stimulated. The method then proceeds to end block **512** and terminates.

Figure 6 is a block diagram depicting an exemplary spectroscopy application that is an exemplary embodiment of the present invention. In the spectroscopy application of Figure 6, a nanosize specimen **600** is located near two SPASER devices **602**, **604**. The SPASER devices are energized as described above and produce SPASER responses that impact the specimen **600**. The specimen may cause the Raman scattering of the SPASER fields. A spectroscopic detector **606** may be used to detect the scattering and to generate an output for the analysis of the scattering. For example, the spectroscopic detector **606** may be configured to identify certain characteristics of the specimen based on the scattering analysis. Because of

the nanostructure of the SPASER device **602, 604**, such specimen identification can be performed on a nanoscale. The applications for use of SPASER devices **602, 604** on the nanoscale are virtually limitless. For example, a single specimen comprising a single molecule, a single cell, or a single virus can be analyzed by one or more SPASER devices. Such a device might be used to detect the presence of a very small amount of a dangerous substance such as an anthrax spore, or a smallpox virus. While conventional devices exist for such detection, the use of a SPASER device in this application would enable significantly earlier detection of significantly smaller specimen sizes, down to a single molecule, virus, spore, or cell. High intensity local fields of a SPASER can be used to modify a molecule or a surface within a nanoscale region. This modification can be photochemical or photophysical (e.g., melting, evaporation, etc.).

Figure 7 is a block diagram depicting an exemplary electronic data storage and retrieval application that is an exemplary embodiment of the present invention. As depicted in Figure 7, a SPASER device **700** can be used to write (e.g., etch) bits of data on the surface of a recording medium **702**. The SPASER device **700** may then be used in conjunction with a read device **704** to read data that has been previously recorded on the surface of the recording medium **702**. The read device **704** may, for example, receive and analyze a radiation field that is emitted by the surface of the recording medium **702**. The nanoscale operating range of the SPASER device **700** enables super-dense recording of data, such that the physical size requirements of the recording medium **702** can be significantly reduced. That is, significantly higher amounts of data can be stored on significantly smaller storage media, by using a SPASER device instead of conventional data writing devices.

Figure 8 is a block diagram of a pseudo-SPASER device that generates both localized electrical and magnetic fields. As described above, the SPASER device of various embodiments of the present invention is characterized by generation of an electric field with little or no magnetic component. The embodiment of Figure 8 uses a resonant medium that has a split-ring configuration that generates both a localized

electric field and a localized magnetic field. However, unlike the nanostructure of the SPASER device, the device of Figure 8 has a much larger structure on the scale of a fraction of a wavelength of light. For example, the diameter of the split-ring resonant medium is one-third wavelength.

Although the present invention has been described in connection with various exemplary embodiments, those of ordinary skill in the art will understand that many modifications can be made thereto within the scope of the claims that follow. Accordingly, it is not intended that the scope of the invention in any way be limited by the above description, but instead be determined entirely by reference to the claims that follow.